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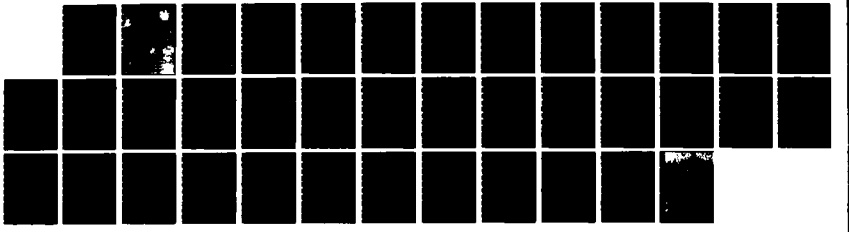

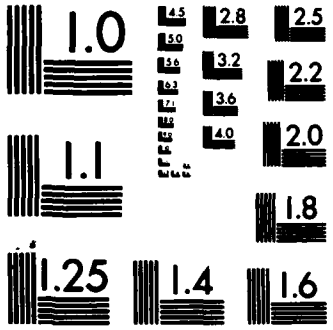

Center for Adhesion

VPI-E-84-12 May 1984

CUMULATIVE CREEP DAMAGE FOR POLYCARBONATE AND POLYSULFONE

by

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CUMULATIVE CREEP DAMAGE

FOR

POLYCARBONATE AND POLYSULFONE

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ABSTRACT

The literature for creep to failure cumulative damage laws are reviewed. Creep to failure tests performed on polycarbonate and polysulfone under single and two step loadings are discussed. A cumulative damage law or modified time fraction rule is developed using a power law for transient creep response as the starting point. Experimental results are approximated well by the new rule. Damage and failure mechanisms associated with the two materials are suggested.

Introduction

During the last two decades, the use of adhesives, polymers and polymer-based composites has become widespread in high performance aerospace and automotive applications. The great potential for composite materials is derived from their high specific strength and modulus properties, improved fatigue resistance, greater flexibility for tailoring material properties to meet design requirements, reduced manufacturing costs and fabrication scrap, and improved dimensional stability due to lower thermal expansion.

Modern adhesives have also received a great deal of attention in recent years partly because of the increased use of polymeric and composite materials in structural components. For these nonmetallic materials, conventional fasteners such as bolts, rivets, and welds are either inefficient or impossible. Penetration methods cause undesirably high stress concentrations and, in the case of composites, sever the fiber reinforcement, drastically lowering the joint efficiency. On the other hand, bonded joints tend to be damage-tolerant due to the high damping behavior of the adhesive layer and less expensive due to lower fabrication cost. For these reasons it is important to study damage induced by fatigue and/or creep for critical design consideration.

Cumulative damage resulting from fatigue, i.e. periodic or time-varying internal stresses created by similar time-varying external loadings, is well known. Less well known is a similar cumulative damage or degradation due to viscoelastically-induced changes in moduli and strength properties of materials. Polymers, adhesives and fiber reinforced plastics (FRP) are particularly susceptible to this effect.

Ample evidence suggests that these materials may suffer delayed failures or ruptures due solely to their viscoelastic or time-dependent material properties [1-6]. Thus, the development of cumulative creep damage laws which may be analogous to well-established cumulative fatigue damage laws is obviously necessary. In fact, the two types of damage laws and hence damage mechanisms may be directly related for polymer-based adhesives and composites.

Consider, for example, a step lap joint as shown in Fig. 1.

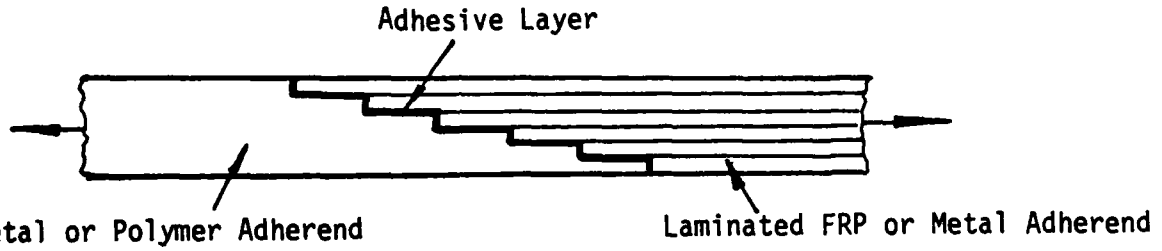

Fig. 1. Step Lap Joint Between Laminated FRP and Metallic or Polymer Adherends.

The joint visualized could be between general laminated composite and metallic adherends or any combination of adherend types for that matter. For a constant stress input, σ_0, a variety of stress distributions and interactions will develop in the adherends and in the adhesive layer. A simple delayed failure, delamination or rupture at an isolated point in the example shown will not cause the joint or structure to cease transmitting load from one adherend to the other. But a failure at one point will cause a step increase in stress at other points, with a corresponding change in failure or rupture time. Again,

this illustrates the need to develop a cumulative creep-damage model for composites and/or adhesive joints even if the remote load is constant. At present there is no universally accepted method for estimating the creep life of a material subjected to multiple stress levels for various periods of time. Several different techniques for making such estimates have been proposed and will be discussed in the next section.

Herein, the cumulative creep damage of polycarbonate and polysulfone is studied as a necessary first step to understanding the more complicated cumulative damage process in polymers, FRP materials and/or adhesively bonded joints. Failure, in the the case of polycarbonate, is defined as yielding. The yielding of polycarbonate is further defined as the elastic-plastic tensile instability point or the point of Lüder's band formation [1]. That is, slip, yield or Lüder's bands occur in polycarbonate at the elastic-plastic tensile instability point as in mild steel. Further, polycarbonate is viscoelastic and, in a creep test, a creep-to-yield or a creep-to-Lüder's band formation will occur [1]. In polysulfone, on the other hand, failure occurs by rupture. In other materials failure might occur simply by excessive deformation. In any event, we feel that an appropriate continuum analytical model for cumulative damage may be applicable to failure regardless of the specific definition of the failure. For example, the same delayed failure (yield) model due to Crochet has been used for yielding in polycarbonate [1], rupture in neat epoxy adhesives [2], and rupture in a sheet molding compound composite SMC-25 [6]. It should be noted again, however, that we feel the present study will have applications to both adhesives and composites even though the materials studied, polycarbonate and polysulfone, are both homogeneous and isotropic.

Review of Literature

Creep is the transient time-dependent component of strain resulting from a constant stress input. Damage may occur at different rates for two different stress levels which leads to a particular creep life at each stress level. The effect of damage accumulation at one stress level on the remaining life at a second stress level represents the aspect of material response defined as cumulative damage.

Robinson [7] originally proposed a creep damage law now often referred to as the "life fraction rule" or the "linear cumulative damage rule", which can be expressed as,

$$\sum_i \frac{t_i}{T_{fi}} = 1 \qquad (1)$$

This equation is valid for a tensile specimen at a constant temperature loaded in such a way that a creep stress σ_1 is input and held constant for the time interval t_1. Next the stress is increased to σ_2, and held constant for the time interval t_2, etc., until final failure. The quantities T_{fi} are the failure (in our case yielding or rupture) times associated with each increment of stress acting alone for the same constant temperature.

For various types of temperature cycles, Miller [8] found the life fraction rule to give quite good predictions for several superalloys. However, later studies [9], involving variable stress tests on superalloys and steels, indicated total lifetimes ranging from 0.36 to 2.08 as calculated by the left hand side of equation 1. Hence, despite the convenience of Robinson's life fraction rule, care and discretion must be exercised in its use.

Randall [10] suggested that the life fraction rule should be useful in estimating cumulative damage only if loads are high enough to produce significant plastic deformation. Also, he suggests that high temperatures should be avoided or such exposure should be of short duration to minimize the effect of solid state reactions. Goldhoff and Woodford [11] suggested that adequate predictions could be made only for small changes in stress.

Because of these limitations of the life fraction rule, other less convenient procedures have been suggested. Lieberman [12] proposed an analogous strain fraction rule which can be written as,

$$\sum_i \frac{\varepsilon_i}{\varepsilon_{fi}} = 1 \qquad (2)$$

where ε_i is the strain under a given stress and temperature and ε_{fi} is the strain at failure under the same stress and temperature. The fraction of the amount of failure lifetime expended is assumed to be equivalent to the amount of creep deformation which occurs for a given stress level regardless of whether it occurs at the start, near the middle, or at the end of a test. Thus, a time interval associated with rapid primary or tertiary creep would consume a large portion of the total lifetime as compared to a time interval associated with secondary creep.

Oding and Burdusky [13] assumed the rate of production of new voids to be proportional to the secondary creep rate $\dot{\varepsilon}$ and the rate of accumulation of voids to be proportional to the period of creep time t raised to an exponent of m. Using this approach they arrived at the law,

$$\sum_i \left[\frac{t_i}{T_{fi}}\right]^m = 1 \qquad (3)$$

Johnson [14] observed that materials which do not exhibit micro-cracking during creep to fracture and obey an effective stress failure criterion adhere to a cumulative time damage rule. In contrast, he suggests that materials exhibiting general micro-cracking during creep and following the maximum principal stress criterion adhere to a cumulative strain damage rule. He therefore modified equation (3) to be applicable to primary and tertiary creep for which cases the creep rate is not a constant but is proportional to time t raised to another exponent μ. The combined expression for primary or tertiary and secondary damage was given as,

$$\sum_i \left[\frac{t_i}{T_{fi}}\right]^{m+\mu} = 1 \qquad (4)$$

In reality, equation (4) is the same as equation (3) but the exponent has a different significance.

Freeman and Voorhees [15] compared predictions by the life and strain fraction rules and found that the errors in the two methods appeared to be in opposite directions. For this reason, they concluded that a rule for consumption of rupture life under variable stress appeared to be somewhere between the two but perhaps closer to the life fraction rule. They suggested that a reasonable prediction could be obtained from the geometric mean of the two approaches,

$$\sum_i \left[\frac{t_i}{T_{fi}} \cdot \frac{\varepsilon_i}{\varepsilon_{fi}}\right]^{1/2} = 1 \qquad (5)$$

Another mixed creep damage rule was proposed by Abo El Ata and

Finnie [16] and can be written as,

$$K \sum_i \frac{t_i}{T_{fi}} + (1-K) \sum_i \frac{(\varepsilon_{max})_i}{(\varepsilon_{max})_{fi}} = 1 \qquad (6)$$

When K ~1, the time fraction rule is recovered and when K ~0, the strain fraction rule is recovered.

A rather involved method to predict the damage accumulation during the creep process in terms of a reduction in material strength was developed by Bui-Quoc [17]. His approach is based upon finding a damage function which is determined, in part, from the concept of a creep endurance limit.

Kargin and Slonimsky [18] have generalized the linear cumulative damage concept to an integral form including temperature effects which is useful for continuously varying external stresses.

$$\int_0^{\tau} \frac{dt}{t_f[\sigma(t),T(t)]} = 1 \qquad (7)$$

Lemaitre and Chaboche [19] developed an equation including both non-linear creep damage accumulation and non-linear fatigue accumulation. The proposed interaction relation is,

$$dD = \left(\frac{\sigma}{A}\right)^r (1-D)^{-k(\sigma)} dt + \left[1 - (1-D)^{(\beta+1)}\right]^{\alpha(\Delta\sigma)} \left[\frac{\Delta\sigma}{M(\bar{\sigma})\ (1-D)}\right]^{\beta} dN \qquad (8)$$

In equation (8), $\bar{\sigma}$ is the mean stress, A, r, β, $k(\sigma)$, $\alpha(\Delta\sigma)$, and $M(\bar{\sigma})$ are temperature dependent material coefficients and functions, and D is a damage term such that $D = D_C + D_F$ in which D_C is creep damage and D_F is fatigue damage.

The cumulative creep damage approaches described above have evolved from similar concepts related to cumulative fatigue damage and are

largely empirical in nature. Futher, these laws have evolved primarily from observations of metallic alloys, usually at elevated temperature. The present effort is an attempt to develop an approach to cumulative creep damage of polymer materials based upon observations for polycarbonate and polysulfone, each of which have very different failure processes, and to relate the model developed directly to the viscoelastic nature of polymeric materials. While the approach we shall take is empirical, we feel that a similar model could be developed as a natural extension to the nonlinear viscoelastic constitutive methods of either Findley [20] or Schapery [21].

Cumulative Damage Model

For most materials other than those which are perfectly linear elastic and perfectly brittle, failure is likely a nonlinear process however it is defined. Our observations on polymers, composites and adhesives suggest that the amount of nonlinearity is both a function of the stress level and the time scale [22,23]. Indeed, various nonlinear theories are in agreement with this observation [20,21]. That is, for high stress levels, nonlinear creep processes occur even over a very short time scale, while for low stress levels, nonlinearities may only be detectable after a very long time.

Unfortunately, for polymers, nonlinear deformation processes may occur for a variety of reasons. For example, as illustrated in Fig. 2, the total strain response for a constant stress input to a four parameter mechanical model is composed of an instantaneous elastic component (free spring), a delayed elastic component (Kelvin element or spring and damper in parallel) and flow (free damper). Each component may

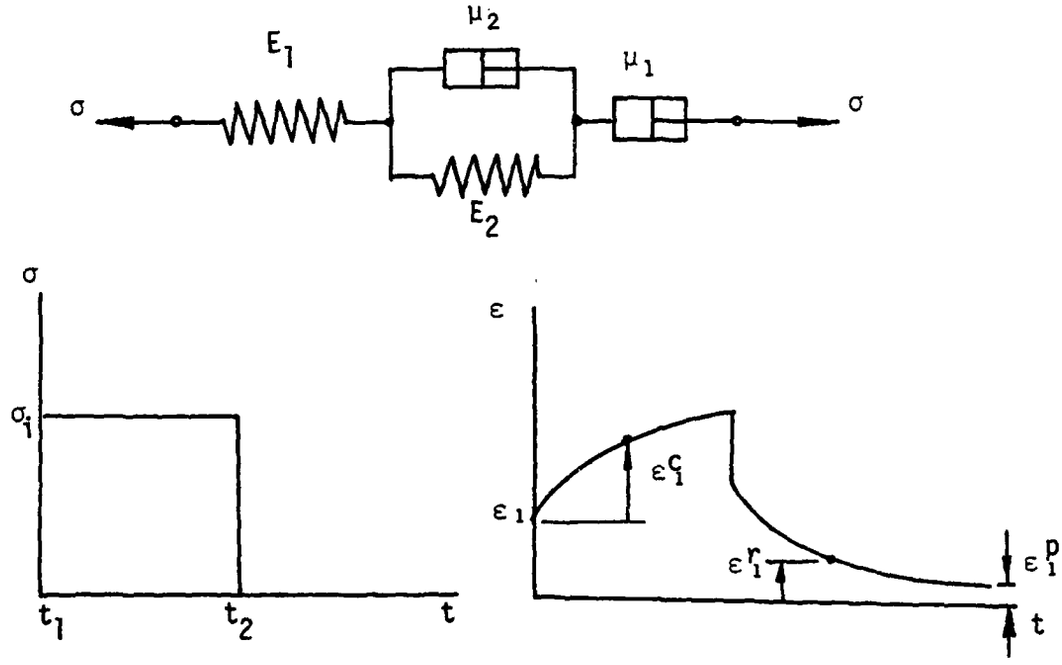

Fig. 2. Creep and Creep Recovery of a Four Parameter Model.

contribute to a nonlinear deformation or strain. Likewise, each component may contribute to the overall accumulation of damage.

Damage is normally assumed to give rise to permanent deformation and the latter is often considered to be evidence of the former. However, the viscous component of the free damper allows for a permanent deformation which may not necessarily be associated with damage production. Thus, damage accumulation for viscoelastic materials is clearly a complicated issue.

In our case, we have performed many creep and creep recovery tests

on polycarbonate and polysulfone. At sufficiently high stress levels or sufficiently long exposure times, we observed a creep-to-failure process which in the case of polycarbonate was defined as yielding and in the case of polysulfone was defined as rupture. For these materials, we found that if the time at which the load was removed, t_2, was very short (about one minute) and if failure had not occurred, almost total recovery was obtained after about one hour. That is, no permanent strain, ε_1^p, was measurable. On the other hand, if the time, t_2, was long (about one hour), total recovery was not obtained after several days. That is, permanent deformation, ε_1^p, occurred. These observations were valid even for creep stress levels as high as ninety percent of the ultimate stress level for which failure would occur upon loading, i.e., $\sigma_1 = 0.9\,\sigma_u$.

For the above reasons and for the range of test conditions that we used, damage for polycarbonate and polysulfone seems to be related to transient strains and not to the instantaneous component. As a result, we will assume that the rate of damage accumulation is proportional to the transient strain such that,

$$\frac{dD_1}{dt} = k_1 \varepsilon_1^c \tag{9}$$

where D_1 is the total damage at any given time under an arbitrary creep stress σ_1 , ε_1^c is the transient creep strain and k_1 is a factor which depends upon stress level.

A power law is often used to represent creep strains [20] and may be written as

$$\varepsilon(t) = \varepsilon_1 + \varepsilon_1^c = \varepsilon_1 + m_1 t^n \tag{10}$$

where ε_1 and m_1 are parameters which are functions of stress level for

a nonlinearly viscoelastic material and n is a constant. Combining (9) and (10) gives,

$$\frac{dD_1}{dt} = k_1 m_1 t^n \tag{11}$$

On the basis of our assumption stated by equations (9) and (11), we propose the empirical relation for total damage at an arbitrary time for a single (but arbitrary) creep stress level to be given as

$$D_1 = K_1 t^N \tag{12}$$

where K_1 is a stress dependent parameter and N is a constant not directly related to n wherein the empiricism arises.

When failure occurs,

$$D_{f1} = K_1 T_{f1}^N \tag{13}$$

where T_{f1} is the creep to failure time under the constant stress σ_1.

For multiple step creep inputs such as those shown in Fig. 3, a nonlinear superposition method is needed in order to ascertain the

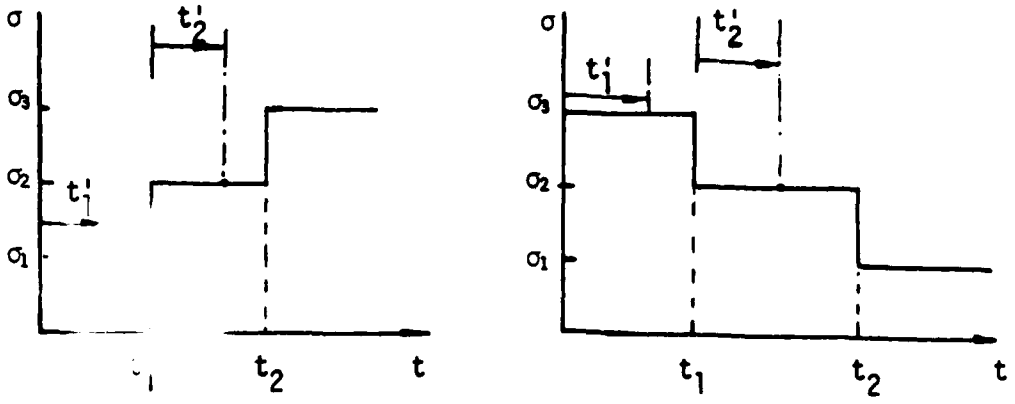

Fig 3. Multiple Step Creep Inputs

strain output. While the Schapery [21] approach is more rigorous, we will use the Findley modified superposition as reported by Dillard, et al. [23]. For a stress input with q steps, the strain output may be written as

$$\varepsilon(t) = \varepsilon_o + \sum_{i=1}^{q} [(m_i - m_{i-1}) (t_i')^n] \tag{14}$$

where $t_i' = t - t_{i-1}$ and zero subscripted quantities are zero.

If we again assume the rate of damage accumulation to be a function of the transient strains only, then the total damage after q steps will be,

$$D_{TOT} = \sum_{i=1}^{q} C_i(m_i - m_{i-1}) (t_i')^N \tag{15}$$

where C_i is a stress dependent factor as is m_i. Another way of expressing the total damage would be,

$$D_{TOT} = \sum_{i=1}^{q} \Delta D_i(t_i') \tag{16}$$

where ΔD_i is the damage which takes place during each step stress increment.

A cumulative damage law for multiple creep step loading can now be obtained by assuming that the sum of the ratios of the damage during each increment to the total damage at rupture for a single creep stress of the same magnitude is equal to one, i.e.,

$$\sum_{i=1}^{q} \frac{\Delta D_i(t_i')}{D_{fi}} = 1 \tag{17}$$

Combining equations (13), (15) and (17) gives,

$$\sum_{i=1}^{q} \frac{C_i(m_i - m_{i-1}) (t_i')^N}{K_i T_{fi}^N} = 1 \tag{18}$$

We finally assume that the coefficients C_i, m_i and K_i can be combined in a simple way such that,

$$\sum_{i=1}^{q} \bar{K}_i \left[\frac{t_i'}{\bar{T}_{fi}}\right]^N = 1 \qquad (19)$$

This last assumption is not very likely as the numerator for each step is a function of all previous history. However, a simple form results which is similar to the time fraction rule given by Oding and Burdusky [13] in Eq. 3.

For a two step loading, Eq. 19 becomes,

$$\left[\frac{t_1'}{\bar{T}_{f1}}\right]^N + \bar{K}_2 \left[\frac{t_2'}{\bar{T}_{f2}}\right]^N = 1 \qquad (20)$$

which, as we shall see, gives a reasonable representation of our experimental data for polycarbonate *and polysulfone*.

Experimental Procedures

The materials used were 0.12 in. (3 mm) sheets of "Tuffak" polycarbonate and 0.125 in. (3.2 mm) sheets of polysulfone (Udel). The latter were made at VPI&SU and were annealed for four hours and cooled at ~2.8°C/hour. The dog bone shaped tensile specimens had a uniform test section length of about 2 in. (50.8 mm) and width of about 0.4 in. (10 mm). Creep tests were performed using a lever type dead weight creep test machine and strains were measured using an extensometer. A minimum of five samples were tested under identical conditions. Mean values were used in all analyses.

Preliminary tests were performed to observe the creep response

under multiple step loadings. These results for a two step loading history are shown in Figs. 4 and 5 for polycarbonate and polysulfone respectively. These curves indicate the type and magnitude of the nonlinearities present in the material. Interestingly, the recovery curves for the high-low stress history are similar to those of Findley [24]. Specifically, for a small decrease in the applied load, the recovery curves first decrease and then increase again. This is evidence of a fluid-type response.

In these preliminary tests, specimens were loaded and allow to creep for only a short time of about one minute and then unloaded. Strains were completely recoverable and under microscopic examination, no evidence of damage was found. On the other hand, if the specimen was allowed to creep for about one-half hour, recovery was not obtained after several days. Upon microscopic examination of these specimens, a large amount of crazing was evident. These observations led to the formulation of the damage law given in the preceding section.

Creep to Failure Observations

Creep-to-failure data for polycarbonate and polysulfone are given in Figs. 6 and 7. As it may be observed a linear relation between stress level and logarithm of time to failure was obtained. These curves were used to find the ultimate stress, σ_u, for instantaneous failure by extrapolation. Also, the creep to failure lifetimes T_{fi} were found from these data.

Obviously, on the basis of Figs. 6 and 7, to perform multiple step creep-to-failure tests for all stress levels would have required an extremely lengthly test program. Rather, we elected to perform all tests

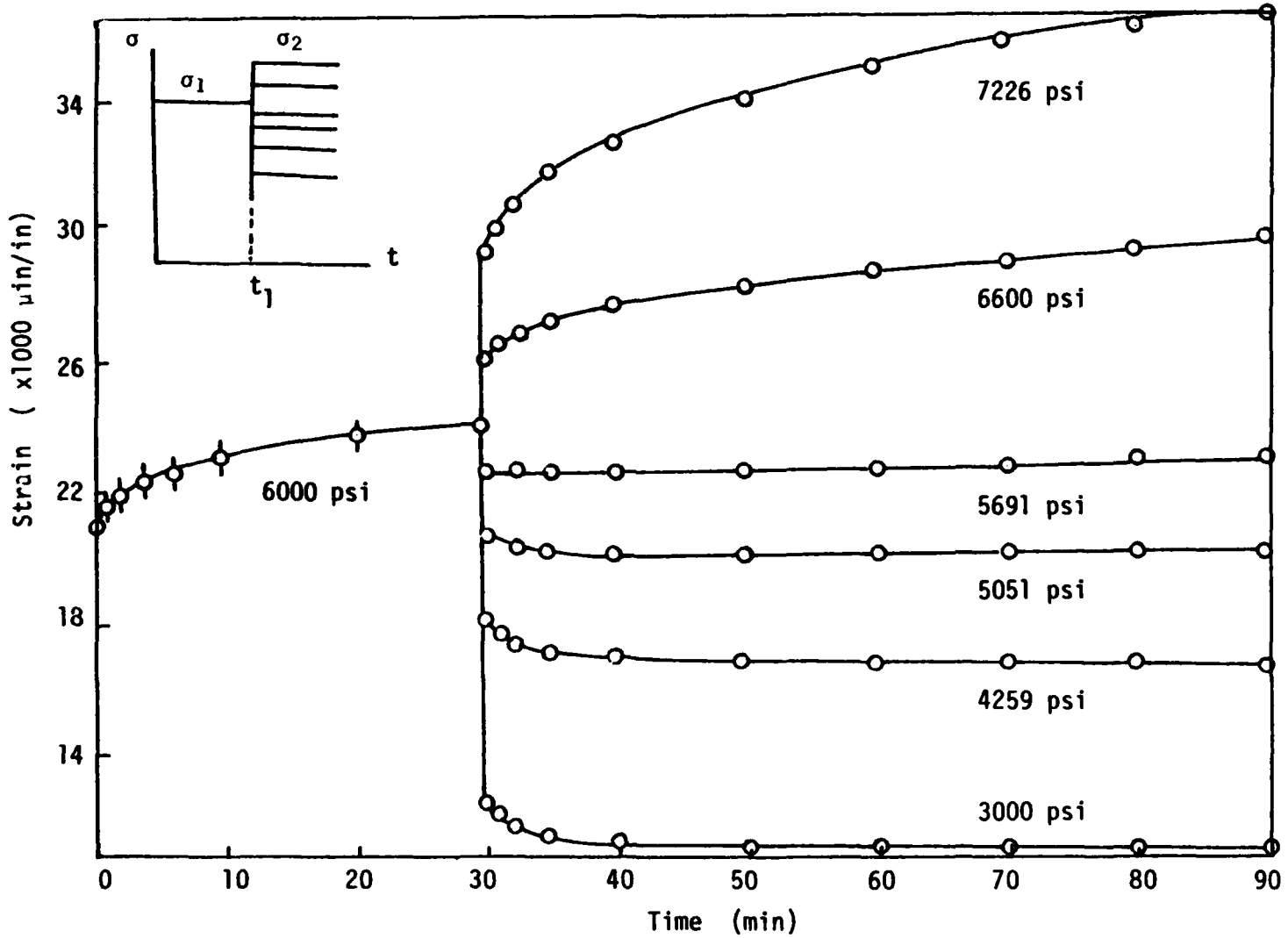

Fig. 4 Creep Curve under Steploading for Polycarbonate at Room Temperature.

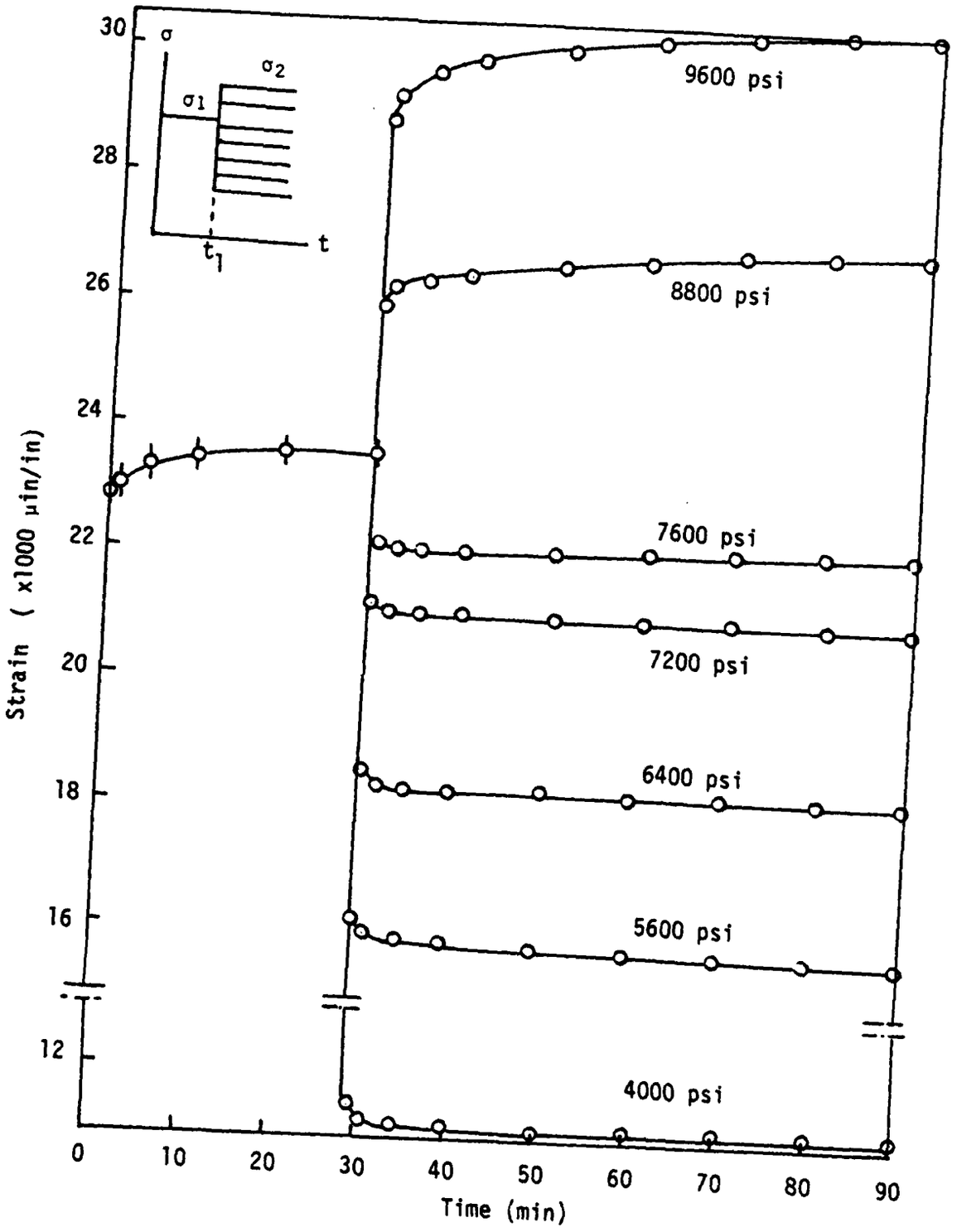

Fig. 5 Creep Curve under Steploading for Polysulfone at Room Temperature.

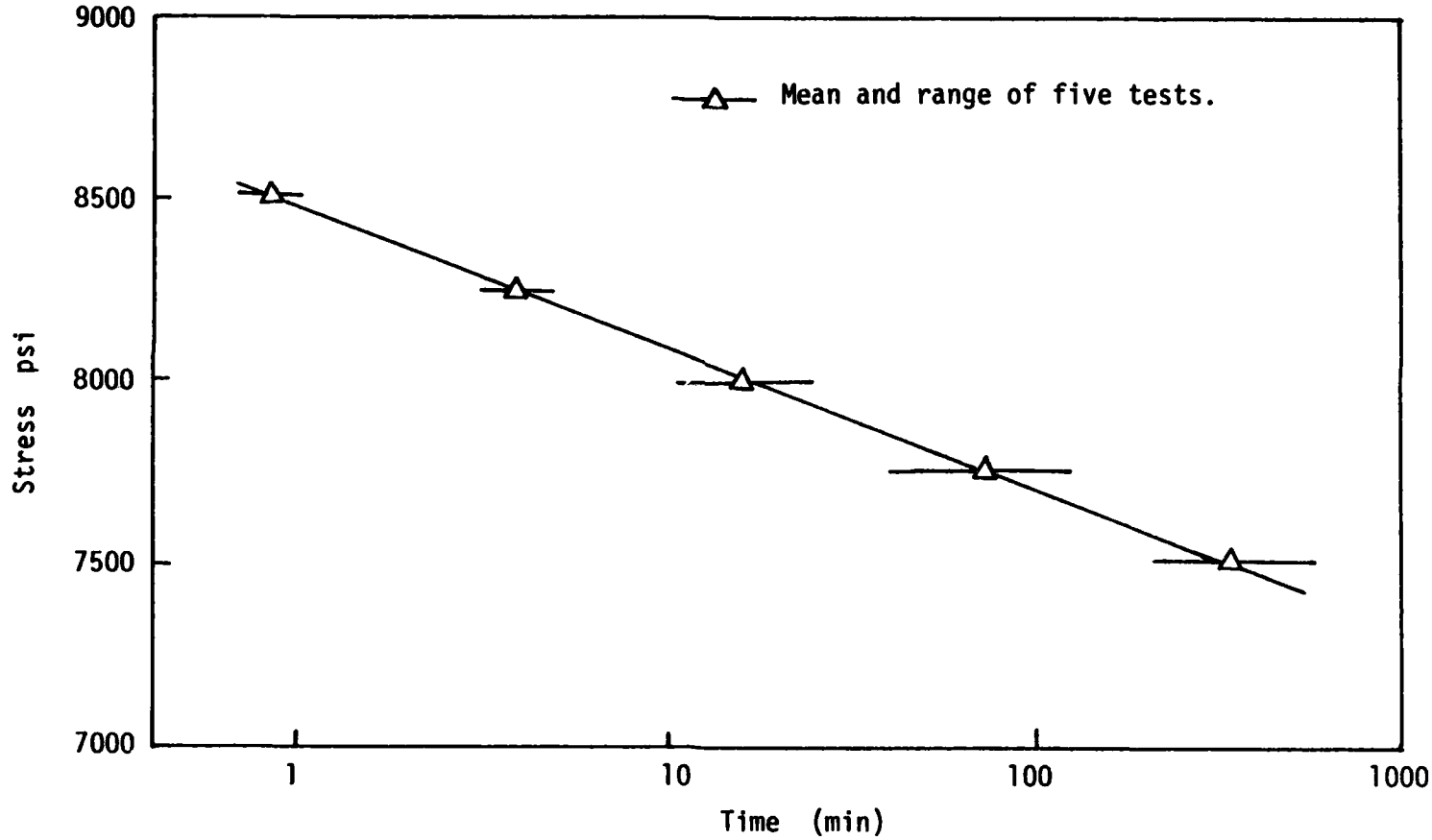

Fig. 6 Creep to Yield Data for Polycarbonate.

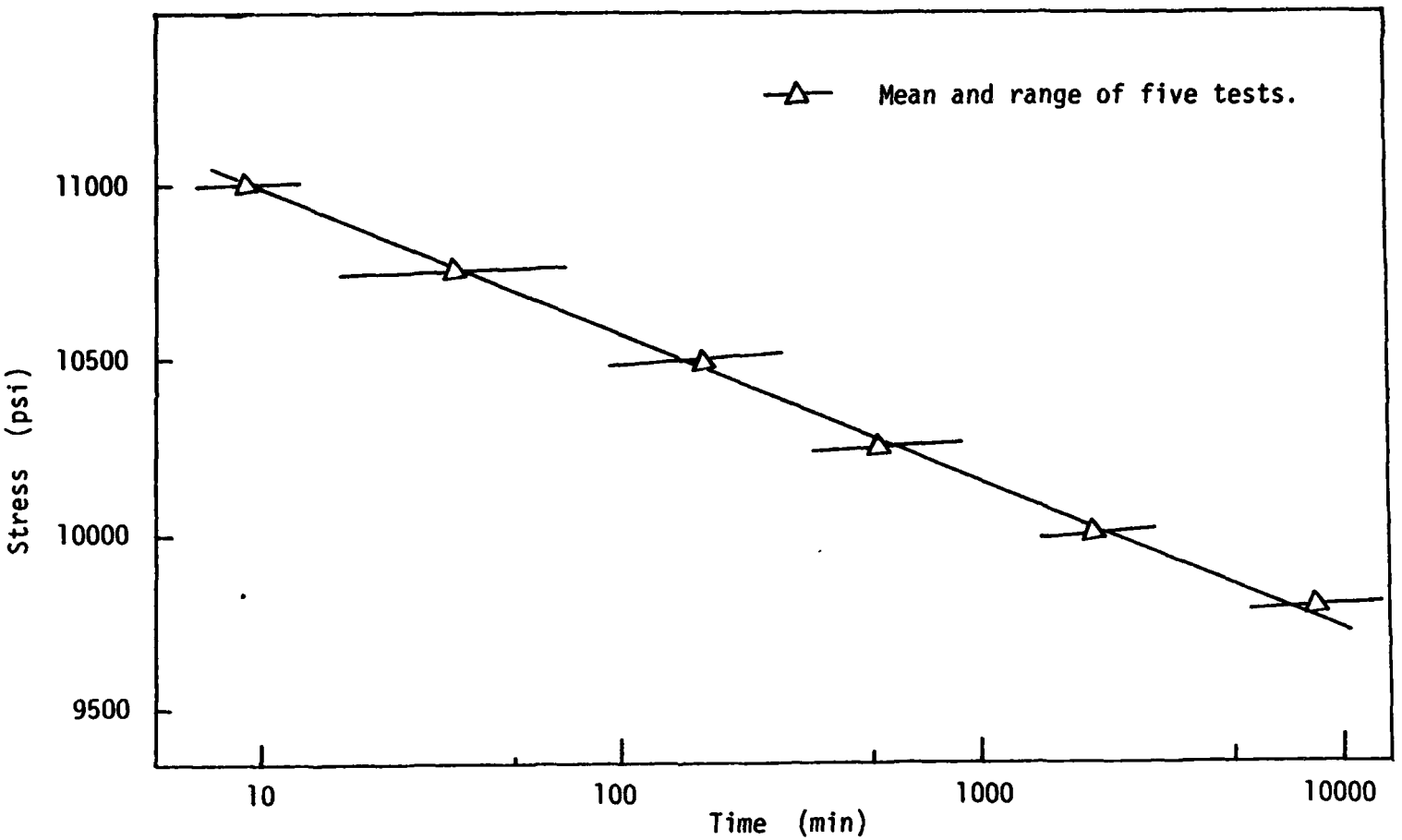

Fig. 7 Creep to Failure Data for Polysulfone.

at very high stress levels. The stress ratios used for the two materials are shown in Table 1.

	Polycarbonate			Polysulfone	
Stress Ratios	High-Low	Low-High Case 1	Low-High Case 2	High-Low	Low-High
σ_1/σ_u	0.904	0.876	0.904	0.947	0.925
σ_2/σ_u	0.876	0.904	0.032	0.925	0.947
$\bar{K}_2$	0.82	0.87	0.73	0.85	0.91
N	0.94	0.76	0.77	1.1	0.86

Table 1

Using these loading schedules, the parameters $\bar{K}_2$ and N were found using Eq. (20). The resulting data and curve fits are shown in Figs. 8-11.

Figs. 8 and 9 show that the cumulative damage curve is shifted to the right of the linear time fraction rule for the high to low step load case. Although polysulfone is subjected to greater non-dimensional stress than polycarbonate, its cumulative curve is further away from the linear case. This tends to indicate that polysulfone is more resistant to cumulative damage. Interestingly, when t_1'/T_1 is less than 0.18 for polycarbonate (Fig. 8) or 0.2 for polysulfone (Fig. 9), t_2'/T_2 values are nearly equal to unity. In other words, there is virtually no damage during the first step loading providing that t_1'/T_1 is less than 0.18 and 0.20 respectively. Our interpretation is that damage does not occur immediately upon loading or during initial creep provided the time ratio for the first step is small. Rather a damage initiation time is required. This phenomenon is consistent with craze formation as

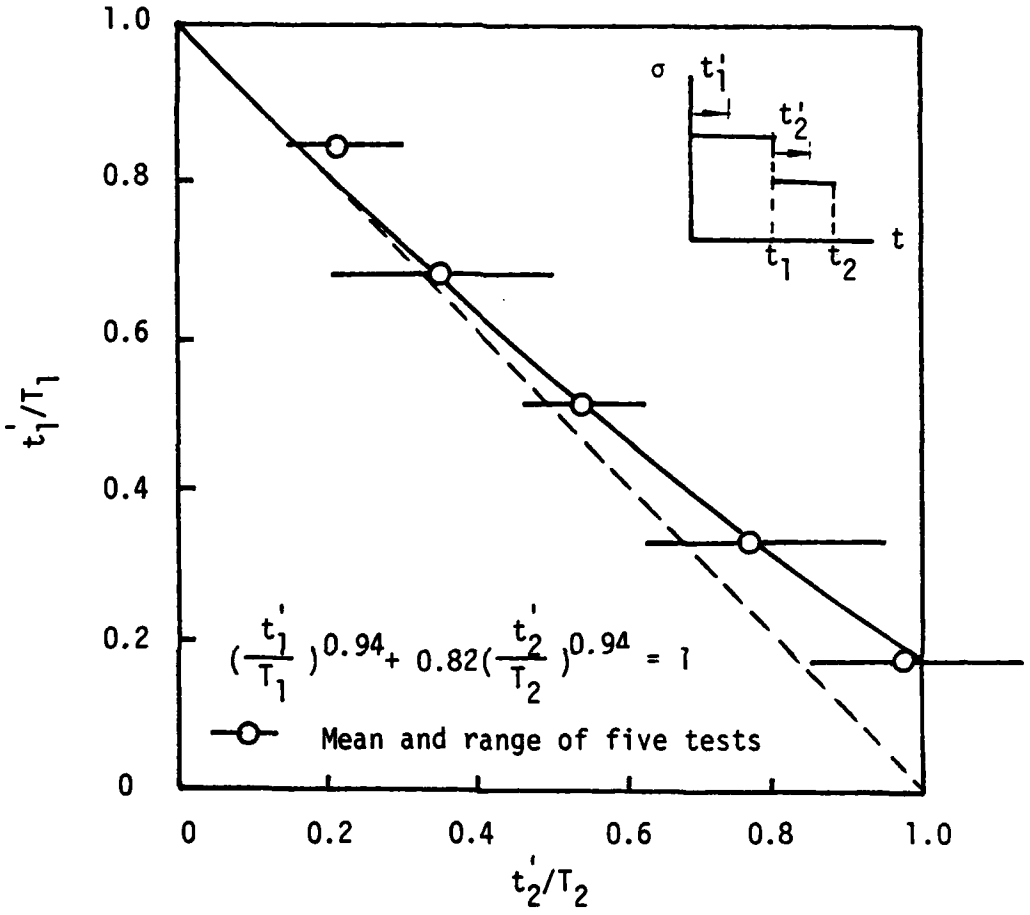

Fig. 8 Cumulative Creep to Yield for Polycarbonate.

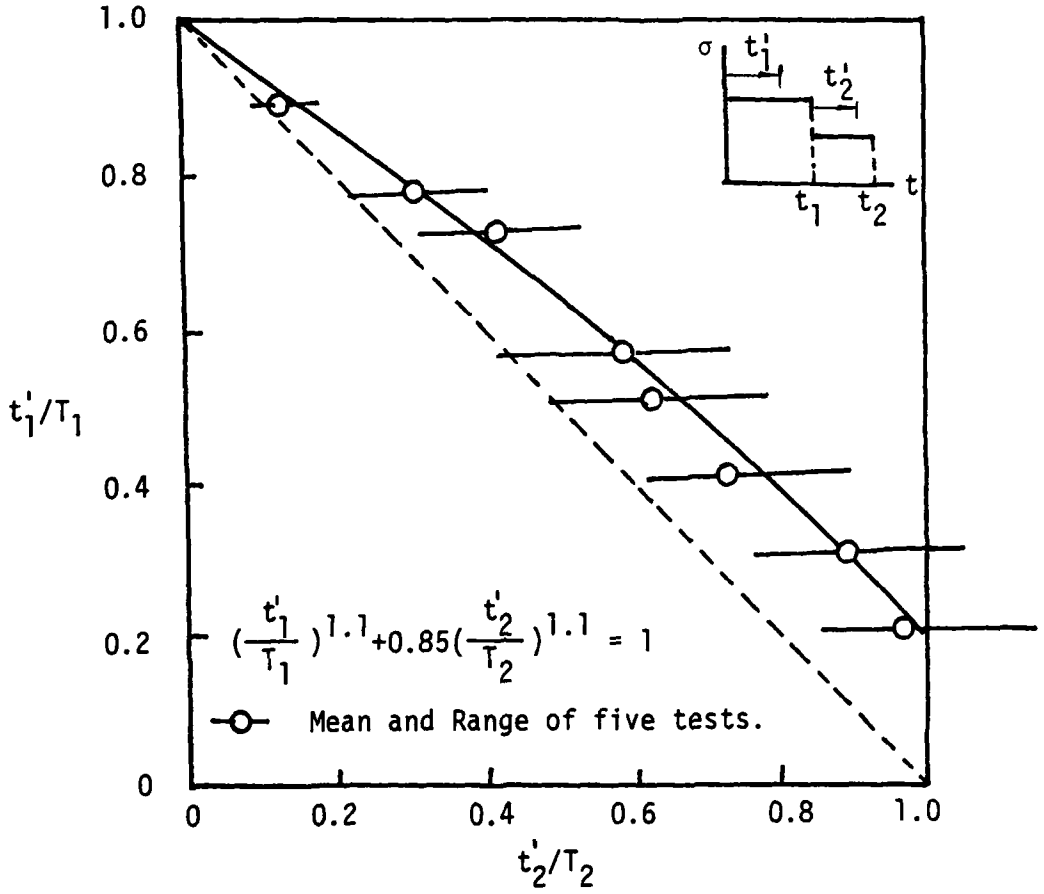

Fig. 9 Cumulative Creep to Rupture for Polysulfone.

reported by others [25-28]. That is, the lower the applied tensile stress or strain, the longer before crazing initiates.

Results for low high creep step loading are shown in Figs. 10 and 11. In all cases, the cumulative damage curves are to the left of the linear time damage rule for large values of t_1'/T_1 and to the right for small values of t_1'/T_1. This trend is much more evident for polycarbonate than polysulfone. In all cases polysulfone appears to be a more damage tolerant material than polycarbonate from the standpoint of failure. However, one must keep in mind the different nature of the failure modes, i.e., yielding for polycarbonate and rupture for polysulfone. Actually, it should be noted that the polysulfone we tested first yielded but was followed almost immediately by rupture. Therefore, mechanisms responsible for damage until the point of failure were not all that different. In this sense, for the small thickness samples used, polycarbonate may be considered more fracture or crack growth tolerant than polysulfone. Thick materials with large constraint effects would likely be very different in their fracture response at the termination of cumulative damage.

Polycarbonate, as shown in Fig. 10 and detailed in Table 1, was tested for two different step stress histories. These results indicate that the stress history plays a major role in the determination of the parameters $\bar{K}_2$ and N as defined by Eq. 20. This gives ample evidence as to why a cumulative damage rule cannot be obtained from nonlinear viscoelastic theories in a simple way.

The results in Fig. 10 for polycarbonate tend to indicate that low step stress levels are very unconservative compared to a linear time

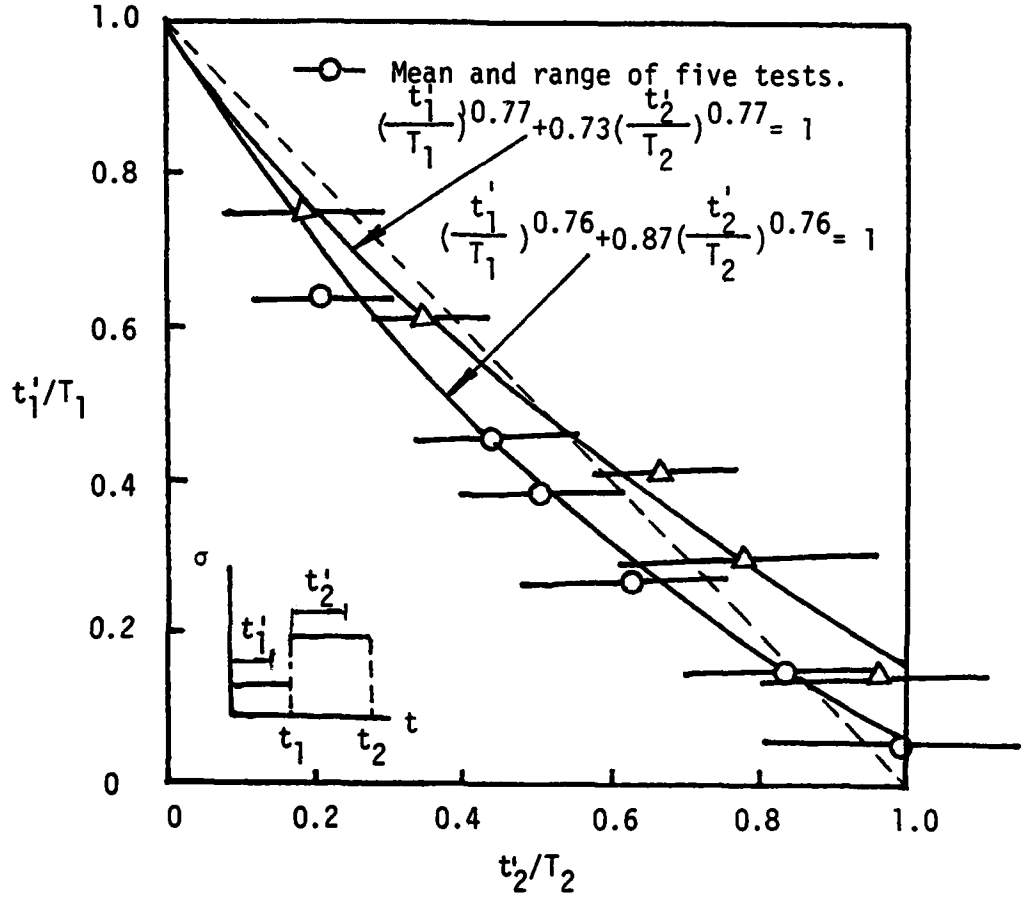

Fig. 10 Cumulative Creep to Yield for Polycarbonate (Triangles and Circles are for two different loading histories).

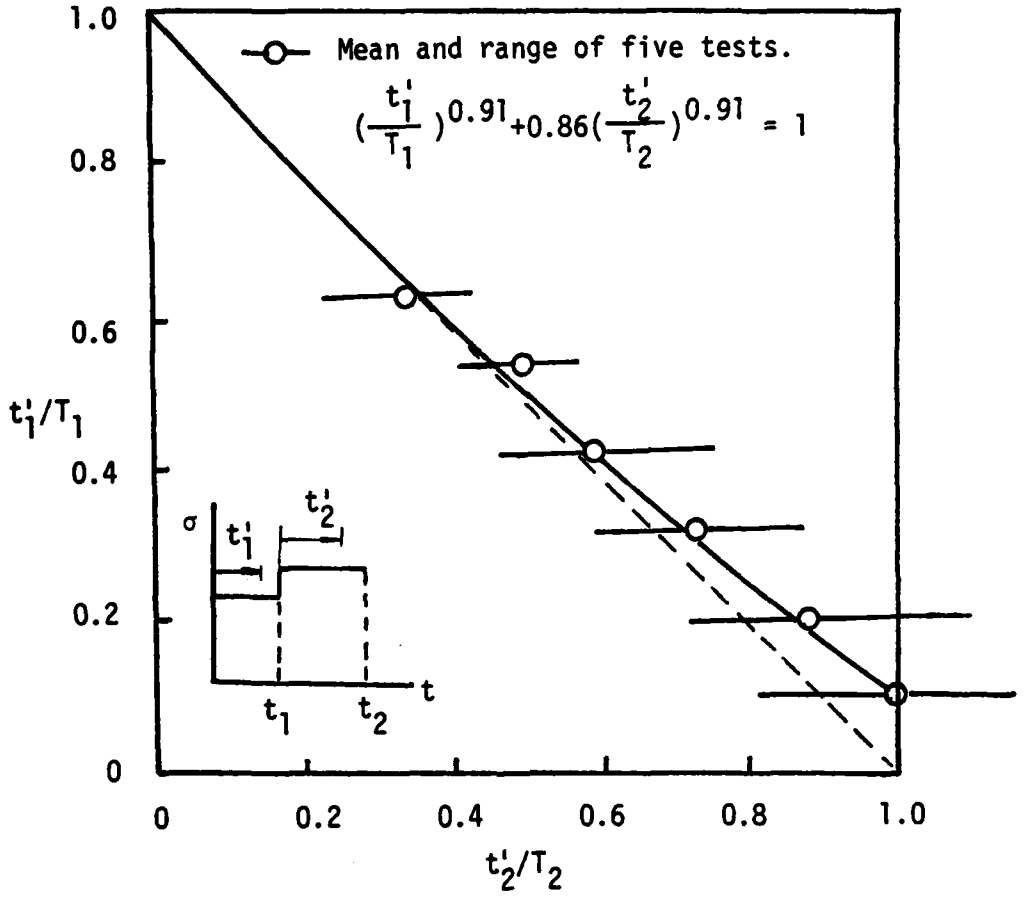

Fig. 11 Cumulative Creep to Rupture for Polysulfone.

damage rule while high stress levels are less unconservative and, in fact, might even be conservative for certain combinations of stress and time ratios.

Again, for small values of t_1'/T_1 , the ratios t_2'/T_2 are nearly unity for low to high stress ratios as they were for high to low stress ratios. Fig. 10 indicates that this initiation period appears to be quite history dependent. It should be noted that initiation time under high-low step loading is greater than under low-high step loading because of the recovery effect during the former and some damage curing may occur. There is also an accelerated damage effect during low-high loading. On other hand, we also found rupture elongation under high-low loading to be a little less than under low-high loading. This effect may be the result of competition between recovery deformation and crazing. Because of the nonuniformity in the material, crazes may have dominated in some regions while recovery deformation dominated in other regions.

Summary and Conclusion

Various empirical approaches to cumulative creep damage were reviewed. On the basis of experimental observations we elected to assume the rate of damage accumlation to be related to the transient strain component of viscoelastic strains in a creep test. Using a power law approximation to creep, which in itself is an empirical formulation, we deduced an empirical damage accumulation model. This model is consistent with Oding and Burdusky's [13] modification of a time fraction rule. Our time fraction rule contains a coefficient $\overline{K}_1$ and an exponent N, each of which are history dependent. We feel that it is in fact this

history dependence which at present precludes the derivation of a life fraction rule from Schapery's nonlinear viscoelastic approach. Perhaps, more research will lead to such a rigorous derivation.

It might be appropriate to note that a simple integration of the rate of damage as given by Eq. (11) will not produce the total damage as given by Eq. (12). In a companion study we have used the Schapery method to characterize the nonlinear viscoelasticity of both polycarbonate and polysulfone [29]. In this study we found values for the power law exponent and it is clear that the exponent N is not simply related thereto. An interesting aside is that we can relate the exponents n and N by using a fractional derivative concept [30]. However, the authors are not able to give physical meaning to a fractional rate of change. Added understanding might be gained using this concept which has been useful in describing other types of viscoelastic data [31].

Experimental creep-to-failure data for polycarbonate and polysulfone has been presented for single and double creep step loading histories. The data has been successfully fitted with our modified life or time fraction rule. Further, the data has given considerable insight into the damage accumulation process for these materials. For example, initial strains do not appear to give rise to damage even for high stress levels for the materials studied. A certain incubation time appears to be necessary for damage and/or craze formation for very low stress levels.

One final point to make is the need for additional studies of this type. We feel that in order to be able to predict service life times for polymer based materials such as adhesives and composites, damage, of necessity, must become a part of the constitutive law. The main dif-

ficulty in the development of such understanding is the need for extensive test programs over a long period of time and the need to have rigorous, but simple, constitutive models to quantify observed behavior. We feel our efforts are a tentative step in this direction.

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The literature for creep to failure cumulative damage laws are reviewed. Creep to failure tests performed on polycarbonate and polysulfone under single and two step loadings are discussed. A cumulative damage law or modified time fraction rule is developed using a power law for transient creep response as the starting point. Experimental results are approximated well by the new rule. Damage and failure mechanisms associated with the two materials are suggested.

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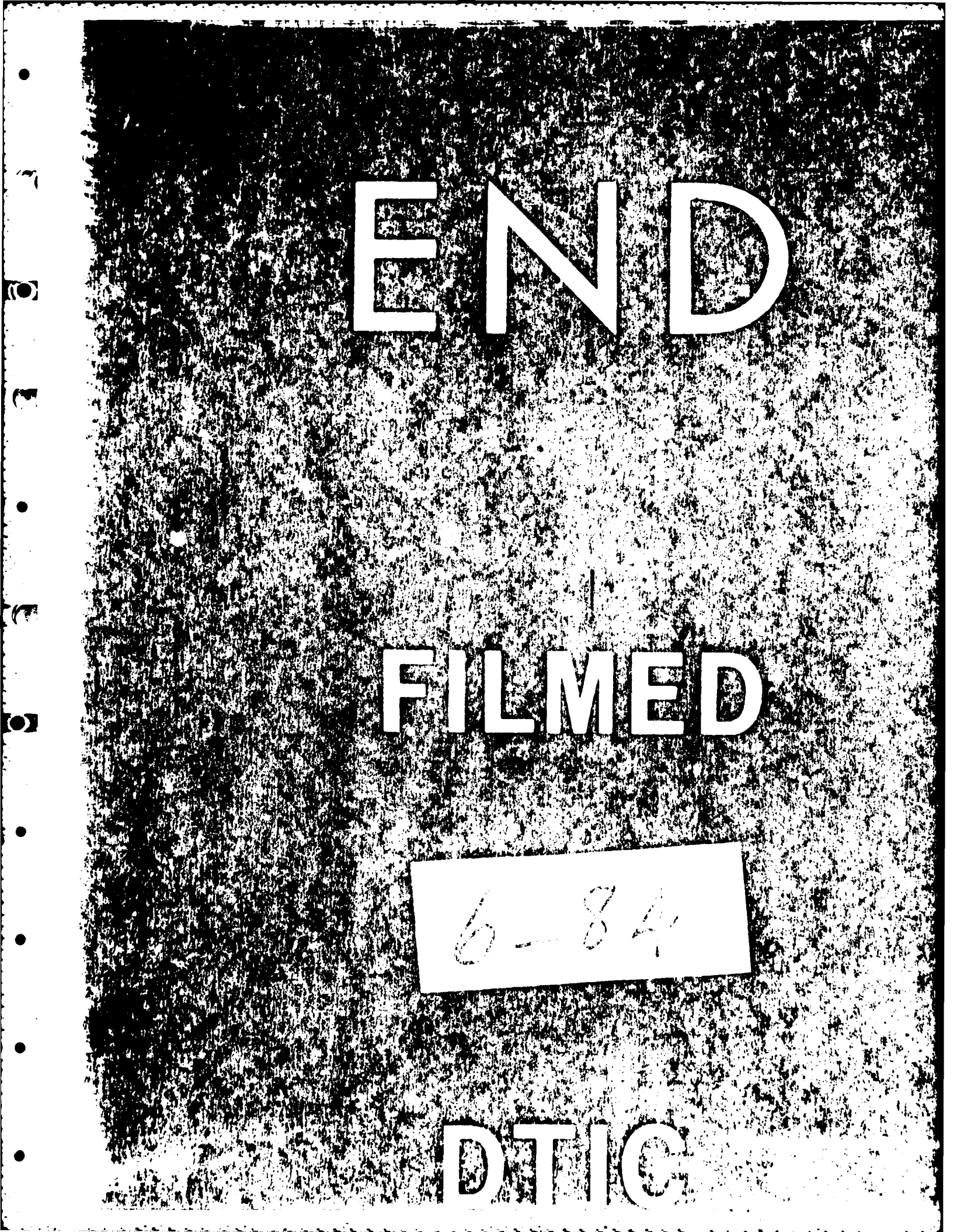
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